

## 意見書

平成16年8月26日

特許庁審査官 平井 裕彰 殿

### 1 事件の表示

平成6年特許願第522432号

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### 4 拒絶理由通知書の日付

平成16年6月1日（発送日）

### 5 理由

[1] 平成16年6月1日（発送日）付けの拒絶理由通知書に鑑み、特許請求の範囲を訂正したので、訂正後の記載に基づいて以下の通り意見を述べます。

#### [2] 本発明の要旨

補正した請求項1に記載の発明（本発明）の要旨は、「基材、化粧層及びその上の保護層からなり、保護層は透明で実質的に熱硬化性樹脂マトリックスと、その中に存在する予備硬化した樹脂粒子及び任意成分としての少なくとも7のモース硬度を有する無機物微粒子とを硬化してなり、予備硬化した樹脂粒子の屈折率は熱硬化性樹脂マトリックスの屈折率と実質的に同じであり、化粧層に樹脂を含浸させると同時に化粧層上に保護層を形成してなる NEMA 耐摩耗性基準を満たす化粧積層体」にあります。

21,000

**Attached Papers**

Specification one

ABSTRACT one

Drawings one

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**Necessity for Proof** Yes

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## 手 続 補 正 書

平成16年8月26日

特許庁審査官 平井 裕彰 殿

### 1 事件の表示

平成6年特許願第522432号

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### 4 補正対象書類名

明細書

### 5 補正対象項目名

特許請求の範囲

### 6 補正の内容

(1) 特許請求の範囲を以下の通り訂正する。

「1. 基材、化粧層及びその上の保護層からなる NEMA 耐摩耗性基準を満たす化粧積層体において、前記保護層は透明で、実質的に熱硬化性樹脂マトリックスと、その中に存在する予備硬化した樹脂粒子及び任意成分としての少なくとも7のモース硬度を有する無機物微粒子とを硬化してなり、前記予備硬化した樹脂粒子の屈折率は前記熱硬化性樹脂マトリックスの屈折率と実質的に同じであり、前記化粧層に前記熱硬化性樹脂を含浸させると同時に前記化粧層上に前記保護層を形成してなることを特徴とする積層体。

I, Ikuzo Tanaka, declare as follows:

1. I am a citizen of Japan residing at 24-5, Mejirodai 4-chome, Hachioji-shi, Tokyo, Japan.

2. To the best of my ability, I translated

Japanese Patent Application No. 2000-157189

from Japanese into English and the attached document is a true and accurate English translation thereof.

3. I further declare that all statements made herein are true, and that all statements made on information and belief are believed to be true; and further that willful false statements and the like are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

Date: August 31, 2004

Ikuzo Tanaka

Ikuzo Tanaka

## SPECIFICATION

### Title of the Invention

#### SEPARATOR FOR ELECTROCHEMICAL FUEL CELL

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### Claims

1. A separator for a fuel cell composed of an aluminum metal plate having flow channels and contact faces that come into contact with electrodes or collectors, wherein an anodized aluminum layer is disposed  
10 on each of said flow channels.
2. The separator for a fuel cell according to claim 1, wherein a conductive film is disposed on said contact faces with electrodes or collectors.
3. The separator for a fuel cell according to claim 2, wherein said  
15 conductive film is made of: a metal selected from the group consisting of Pt, Au, Pd, Ru, Rh, Ir and Ag, or alloys composed thereof; carbon; or a conductive carbide.
4. The separator for a fuel cell according to any one of claims 1-3, wherein said anodized aluminum layer has a porosity of less than 5 %.
- 20 5. The separator for a fuel cell according to any one of claims 1-4, wherein said anodized aluminum layer has a thickness of 5 to 50  $\mu\text{m}$ .
6. The separator for a fuel cell according to any one of claims 1-3, wherein said anodized aluminum layer is composed of a dense anodized aluminum layer having a porosity of less than 5 %, and a porous anodized  
25 aluminum layer having a porosity of 10 % or more provided on said dense anodized aluminum layer.
7. The separator for a fuel cell according to any one of claims 1-6, wherein a corner portion formed between surfaces of said flow channel is

## Japanese Patent Application No. 2000-157189

in shape of a curved surface having a curvature radius of 0.5 mm or more.

8. The separator for a fuel cell according to any one of claims 1-7,  
wherein a corner portion formed between a side surface of said flow  
channel and said contact face is in shape of a curved surface having a  
5 curvature radius of 0.3 mm or more.
9. The separator for a fuel cell according to any one of claims 1-8,  
wherein said aluminum metal plate has a purity of 99.5 % or more.

## DETAILED EXPLANATION OF THE INVENTION

[0001]

### Field of Invention

The present invention relates to a separator for a fuel cell,  
5 particularly to a separator applicable to an on-vehicle fuel cell for an automobile.

[0002]

Fuel cells have attracted much attention as next generation electric generators because they are excellent in energy conversion efficiency and  
10 discharge no harmful or toxic substances. In particular, fuel cells using a polymer electrolyte membrane that work at 150 °C or less have been studied actively and expected to be made practicable a few years later. These fuel cells can work at a relatively low temperature, be high in output density of electric generation, and be miniaturized, thereby suitable for  
15 domestic or on-vehicle fuel cells.

[0003]

In general, the fuel cell using a polymer electrolyte membrane produced by disposing a fuel electrode and an oxygen electrode (air electrode) on both sides of a solid polymer electrolyte membrane to prepare  
20 a unit cell (cell), and by stacking a plurality of the cells with each other through plate-shaped separators having flow channels for supplying fuel gas and air. Used as the solid polymer electrolyte membrane is generally an ion exchange membrane of a fluororesin having a sulfonic acid group, etc., and the fuel electrode and the oxygen electrode are generally made of  
25 carbon black in which a water repellent material such as PTFE and a catalyst such as a noble metal fine particle are dispersed, etc. When a hydrogen-oxygen fuel cell works, protons provided by oxidation of hydrogen gas penetrates into an electrolyte and combines with water



molecules therein to form  $\text{H}_3\text{O}^+$ , and the resultant  $\text{H}_3\text{O}^+$  moves to a cathode. In the cathode, oxygen supplied through flow channels obtains electrons provided by oxidation of hydrogen gas, and combines with protons in the electrolyte to provide water. These processes are repeated to obtain  
5 electric energy continuously. Although the theoretical electromotive force of the hydrogen-oxygen fuel cell is 1.2 V, the actual output voltage is approximately 0.6 to 0.8 V because of voltage drop due to polarization of the electrode, crossover of the reaction gas where the fuel gas leaks to the cathode through the electrolyte, contact resistance of the electrode and the  
10 collector, etc. Accordingly, to obtain the practical output voltage, it is necessary to stack dozens of cells through the separators and connect the stacked cells in series.

[0004]

As known from the above-mentioned principle of electric  
15 generation,  $\text{H}^+$  exists in the electrolyte in large amounts, so that the inside of the electrolyte and the neighborhood of the electrodes where water or water vapor exists in large amounts are strongly acidic. Further, although oxygen combines with  $\text{H}^+$  to provide water in the cathode, hydrogen peroxide may be often provided depending on the working conditions.  
20 The separators are used under such circumstances, thereby required to have high chemical stability and electrochemical stability (corrosion resistance) in addition to electric conductivity and air-tightness.

[0005]

Most of the conventional separators have been produced by  
25 machining a graphite plate. Although the graphite separators are low in electric resistance and high in corrosion resistance, they are poor in mechanical strength and high in machining cost. Thus, it is difficult to utilize the conventional graphite separators as the separator for the

on-vehicle fuel cells, which is required to be high in the mechanical strength and low in the machining cost. Recently proposed is a separator that is produced by mixing graphite powder with a resin, and by injection-molding and baking the resulting mixture, however, there is a  
5 problem that it is low in density to be poor in the air-tightness. Although the density can be increased by impregnating the separator with a resin and by burning the resultant again to carbonize it, this results in complicated manufacturing processes. In addition, thus-produced separator has the contact electric resistance several times higher than that of the conventional  
10 graphite separator, whereby the fuel cell using this separator inevitably lowered with respect to the output voltage.

[0006]

Metal separators have also been studied in addition to the graphite separators. The metal separators are low in bulk electric resistance, high  
15 in air-tightness and mechanical strength, and can be easily produced with reduced working cost. Further, the metal separators can be thinned and miniaturized with ease, and the weight of a fuel cell using the metal separator can be reduced if the metal separator is made of a light metal such as aluminum. However, there is a problem that the metal material is  
20 liable to corrode, in particular, it has been known that aluminum exhibits extremely high corrosion rate (R. L. Rorup et al., Mater. Res. Soc. Symp. Proc., 393 (1995), etc.). Further, there is a fear that metal ions generated by corrosion of the metal material penetrate into the electrolyte membrane to reduce the ion-conductivity thereof.

25 [0007]

Japanese Patent Laid-Open No. 11-162478 has disclosed that the corrosion resistance of the metal separator can be improved by plating entire surfaces thereof with a noble metal. Although this metal separator

plated with a noble metal sufficiently acts as a separator, it necessitates high production cost to be far from practicable. To reduce the production cost, a noble metal layer should be thinned. However, when a thinner noble metal layer is disposed by wet plating methods, the resultant layer has fine pinholes that cause corrosion of the metal separator. On the other hand, dry plating methods such as vapor deposition methods and sputtering methods are poor in efficiency of producing the thinner noble metal layer, and the resultant layer is poor in uniformity.

[0008]

#### 10 **Problems to be Solved by the Invention**

Accordingly, an object of the present invention is to provide a separator composed of an aluminum metal plate for a fuel cell excellent in corrosion resistance.

[0009]

#### 15 **Means for Solving the Problems**

As a result of intense research in view of the above object, the inventors have found that corrosion resistance of a separator made of aluminum for a fuel cell can be improved by disposing an anodized aluminum layer on a small part thereof. The present invention has been accomplished by the finding.

[0010]

Thus, a separator for a fuel cell of the present invention is composed of an aluminum metal plate having flow channels and contact faces that come into contact with electrodes or collectors, wherein an anodized aluminum layer is disposed on each of said flow channels.

[0011]

Also, the separator of the present invention is to exhibit much more excellent corrosion resistance by satisfying the following nine conditions.

- (1) It is preferable that a conductive film is disposed on the contact faces with electrodes or collectors.
- (2) It is preferable that the conductive film is made of: a metal selected from the group consisting of Pt, Au, Pd, Ru, Rh, Ir and Ag, or alloys composed thereof; carbon; or a conductive carbide.
- (3) It is preferable that an anodized aluminum layer has a porosity of less than 5 %.
- (4) It is preferable that the anodized aluminum layer has a thickness of 5 to 50  $\mu\text{m}$ .
- 10 (5) It is preferable that the anodized aluminum layer is composed of a dense anodized aluminum layer having a porosity of less than 5 %, and a porous anodized aluminum layer having a porosity of 10 % or more provided on the dense anodized aluminum layer.
- (6) It is preferable that a corner portion formed between surfaces of the flow channel is in shape of a curved surface having a curvature radius of 0.5 mm or more.
- 15 (7) It is preferable that a corner portion formed between a side surface of the flow channel and the contact face is in shape of a curved surface having a curvature radius of 0.3 mm or more.
- 20 (8) It is preferable that the aluminum metal plate has a purity of 99.5 % or more.

[0012]

### **Mode for Carrying out the Invention**

A separator for a fuel cell of the present invention is composed of an aluminum metal plate and there is disposed an anodized aluminum layer on a small part thereof. The use of aluminum metal as a substrate makes it possible to be light in weight and exhibit improved electrical conductivity and vibration resistant. The separator of the present invention can be

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suitably used in various fuel cells, particularly in on-vehicle fuel cells for automobiles. While the separator of the present invention will be explained in detail below with reference to Figs. 1 to 4, the present invention is not restricted thereto and any modifications may be made  
5 therein unless they deviate from the scope of the present invention.

[0013]

Fig. 1 is a fragmentary schematic view showing an example of a fuel cell using a separator of the present invention. The fuel cell of Fig. 1 is constituted by stacking cells 1 with each other through a separator 5,  
10 each cell 1 being prepared by disposing an anode 3 and a cathode 4 on both surfaces of a solid electrolyte 2. Both ends of the stack are generally connected to an external circuit (not shown).

[0014]

A separator of the present invention has contact faces with  
15 electrodes or collectors. The contact face may have any shape if only the contact face can suitably come in contact with a carbon paper, carbon cloth, etc. of the electrodes or the primary collectors in the fuel cell, and The shape of the contact face is not restricted by Fig. 1.

[0015]

20 As shown in Fig. 1, the separator 5 has flow channels 8 and 9. A fuel gas is supplied through passages each formed by the flow channel 9 and the anode 3, and an oxidizing gas is supplied through passages each formed by the flow channel 8 and the cathode 4. The flow channels 8 and 9 may be formed into a predetermined pattern by machining, pressing,  
25 precision casting, chemical polishing (etching), electro-polishing, etc. Although the shape of the flow channel 8 in Fig. 1 is in C-shape, the flow channel has no particular limitation on the shape thereof if only the shape is such that the flow channel can form the above passage. The flow channel

preferably has such a shape that the flow resistance of the reaction gas is reduced and that the electric generation efficiency is increased.

Generally, the depth of the flow channel is preferably 0.2 to 2 mm, and the width thereof is preferably 0.5 to 5 mm.

5 [0016]

In the present invention, to secure the corrosion resistance of the separator, on each of the surfaces of the flow channels mentioned above that do not come into contact with electrodes, etc. is disposed an anodized aluminum layer 6 that is chemically and physically stable. The anodized  
10 aluminum layer may be disposed by an anode oxidation method, etc. For example, the metal plate may be subjected to electrolysis using an electrolytic solution of an aqueous solution comprising oxalic acid, sulfuric acid, chromic acid, etc., to provide a  $\gamma$ -alumina layer on the surface thereof.

[0017]

15 A hard, dense anodized aluminum layer may be disposed by appropriately selecting anode oxidation conditions, thereby further increasing the corrosion resistance of the separator. The separator is preferably treated with boiling water or water vapor after the anode oxidation to close fine pores of the anodized aluminum layer, thereby  
20 further improving the corrosion resistance thereof. The anodized aluminum layer has preferably a porosity of less than 5 %. In this case, a thickness of the anodized aluminum layer is preferably 5 to 50  $\mu\text{m}$ , more preferably 10 to 30  $\mu\text{m}$ .

[0018]

25 The anodized aluminum layer may have fine vertical pores, or may be composed of a sponge-like porous layer. As shown in Fig. 2, in the separator of the present invention, it is also preferred that the anodized aluminum layer 6 is composed of a dense anodized aluminum layer 11 and

a porous anodized aluminum layer 10 provided thereon to maintain the corrosion resistance for a long period. The porosity of the dense anodized aluminum layer is preferably less than 5 %, and the porosity of the porous anodized aluminum layer is preferably 10 % or more. In this case, the  
5 dense anodized aluminum layer preferably has the thickness of 2 to 30  $\mu\text{m}$ , the porous anodized aluminum layer preferably has the thickness of 5 to 50  $\mu\text{m}$ . The anodized aluminum layer is often swelled and strained by water vapor to be cracked or peeled off, whereby the corrosion resistance thereof seems reduced. In the separator of the present invention, the strain may  
10 be moderated by the porous anodized aluminum layer mentioned above.

[0019]

In the case where the metal plate of aluminum contains a large amount of impurities, the anodized aluminum layer disposed thereon is poor in uniformity and low in density. In such a case, the fine pores of the  
15 anodized aluminum layer are not sufficiently closed by the densification treatment using boiling water or water vapor. Accordingly, the purity of aluminum composing the separator of the present invention is preferably 99.5 % or more, more preferably 99.9 % or more. Further, although there is no particular limitation on the thickness of the metal plate, it is  
20 preferably 0.5 to 3 mm when the separator is used for the on-vehicle fuel cell.

[0020]

In the present invention, as shown in Fig. 1, the conductive film 7 is preferably disposed on each of the contact faces that come into contact with  
25 electrodes or collectors. Thus, the whole surface of the separator according to the present invention is preferably covered with the combination of the non-conductive anodized aluminum layer and the conductive film.

[0021]

The conductive film is preferably made of an electrically conductive material having a high corrosion resistance, and particularly preferably made of: a metal selected from the group consisting of Pt, Au, Pd, Ru, Rh, Ir and Ag, or alloys composed thereof; carbon; or a conductive carbide. The conductive film of a noble metal such as Au, Ag, Pt, Pd, etc. is low in the contact resistance and excellent in the corrosion resistance. The conductive film of carbon is preferably a graphite film provided by CVD, a DLC film (diamond-like carbon film), etc. Further, the conductive film may be disposed by applying a graphite powder containing a water repellent agent to the contact face. The conductive film of carbon can conformably come into contact with the electrode made of carbon black and a slight amount of Pt. The conductive carbide used for the conductive film is preferably silicon carbide, niobium carbide, tungsten carbide, etc. The conductive film of the conductive carbide has a low contact resistance, and excellent corrosion resistance and oxidation resistance, to also act as a protective film of the separator.

[0022]

The conductive film may be disposed by a sputtering method, an electroplating method, a wet plating method, a CVD method, etc. The conductive film preferably has a thickness of 0.01 to 5  $\mu\text{m}$ . When the thickness is less than 0.01  $\mu\text{m}$ , the conductive film is poor in strength to be unstable. On the other hand, the conductive film having a thickness of more than 5  $\mu\text{m}$  results in high cost.

[0023]

When the separators are integrated into the fuel cell, they are generally compressed in a stacking direction under a surface pressure of approximately 1 to 10  $\text{kg}/\text{cm}^2$ , to improve the air-tightness of the fuel cell,



and to sufficiently make the contact faces come into contact with the electrodes or the collectors. The fuel cell generally works at approximately 80 to 120 °C, and the separator in the fuel cell may be deformed by thermal expansion at such a temperature. In particular, there is a case where the anodized aluminum layer is cracked or damaged by deformation of the separator at a portion to which a large stress is applied, thereby resulting sometimes in remarkable decrease in the corrosion resistance. Thus, in the separator of the present invention, a corner portion formed between surfaces of the flow channel, which is shown as R in Fig. 3, is preferably in shape of a curved surface to relax the stress. The curvature radius of the curved surface is preferably 0.5 mm or more, more preferably 1.0 mm or more.

[0024]

To a boundary between the anodized aluminum layer and the conductive film is often intensively applied the stress. Further, the anodized aluminum layer and the conductive film grow vertically to the metal plate, not laterally, so that the anodized aluminum layer is easily damaged at the boundary. Thus, in the separator of the present invention, a corner portion formed between a side surface of the flow channel and the contact face, which is shown as R in Fig. 4, is preferably in shape of a curved surface. The curvature radius of the curved surface is preferably 0.3 mm or more, more preferably 0.5 mm or more. In the separator having such a curved surface, the anodized aluminum layer is sufficiently prevented from swelling and straining.

[0025]

## EXAMPLES

The present invention will be described in more detail referring to examples below without intention of restricting the scope of the invention

thereto.

[0026]

Example 1 and Comparative Examples 1-3

An aluminum metal plate of 1 mm × 150 mm × 150 mm in size  
5 having a purity of 99.6 % was press-worked to form flow channels having  
a depth of 1.0 mm and a width of 3.0 mm thereon, so that a separator base  
was prepared. This separator base was anodized in an oxalic acid aqueous  
solution, immersed in boiling water for 30 minutes, and dried to dispose an  
anodized aluminum layer having a thickness of 12 μm thereon. Then,  
10 electrode contact faces of the anodized separator base were lapped and  
washed, to improve the flatness thereof and remove the anodized aluminum  
layer thereon. Subsequently, each of the electrode contact faces was  
sputtered with Au at a separator base temperature of 200 °C under pure  
argon gas of 5 mTorr to dispose a conductive film having a thickness of  
15 approximately 1 μm, so that a separator was produced.

[0027]

To 100 parts by weight of carbon black was added 15 parts by  
weight of a Pt paste (Pt-content: 90 weight %), and to this was further  
added 15 parts by weight of Teflon particles (average particle size: 0.2 μm)  
20 as a water repellent agent, to prepare a paste for electrodes. The paste was  
applied onto a solid electrolyte membrane of a proton conductive polymer  
(Nafion) and dried. This was put between carbon cloths, and further  
sandwiched between two separators mentioned above, to produce a fuel  
cell (unit cell) Example 1 (hereinafter referred to EX1) using separators  
25 according to the present invention. Incidentally, the separators were  
compressed in a stacking direction under a surface pressure of 10 kg/cm<sup>2</sup>.

[0028]

Comparative fuel cells Comparative Example 1 (hereinafter referred

to CE1) and CE2 were produced in the same manner as the fuel cell EX1 except for using graphite separators or stainless separators (SUS304), respectively. Further, a comparative fuel cell CE3 was produced in the same manner as the fuel cell EX1 except for disposing no anodized aluminum layer on the separator base. Incidentally, on each of electrode contact faces of the separators used in the comparative fuel cells CE2 and CE3 were disposed conductive films of Au, similarly to the fuel cell EX1.

[0029]

To each of the resulting fuel cells (unit cells) EX1 and CE1 to CE3 was supplied a humidified, simulated fuel gas containing 70 volume % of H<sub>2</sub>, 15 volume % of CO<sub>2</sub> and 15 volume % of H<sub>2</sub>O through the flow channels on the anode side, and supplied air as an oxidizing agent through the flow channels on the cathode side, so that each of the fuel cells EX1 and CE1 to CE3 was evaluated with respect to the stability of electric generation. Material used for separator, initial electric generation voltage, electric generation voltage after working for 10 days, and condition of separator after working for 10 days of each fuel cell were shown in Table 1.

[0030]

Table 1

Fuel cell	Material used for separator	Initial electric generation voltage (V)	Electric generation voltage after working for 10 days (V)	Condition of separator after working for 10 days
EX1	Aluminum (with anodized aluminum layer)	0.84	0.80	a*
CE1	Graphite	0.85	0.83	—
CE2	SUS304	0.80	0.68	c*
CE3	Aluminum (without anodized aluminum layer)	0.80	0.20	d*

a\*: Corrosion was not observed.

c\*: Corrosion was observed in part of flow channels.

d\*: Corrosion was observed in whole surface of flow channels.

[0031]

As shown in Table 1, the separator of the present invention having the anodized aluminum layer was excellent in the corrosion resistance,

whereby the fuel cell EX1 using the separator exhibited high stability of electric generation.

[0032]

Example 2 and Comparative Example 4

5           Fuel cells EX2a to EX2r and CE4 using the separators of the present invention were produced in the same manner as the above-mentioned fuel cell EX1 except for using materials shown in Table 2 instead of Au for the conductive film, respectively. Here, the conductive films of the fuel cells EX2q and EX2r were each disposed by sputtering carbon (fuel cell EX2q)  
10 or conductive carbide SiC (fuel cell EX2r) to the electrode contact faces under argon gas of 30 mTorr while using a target thereof.

          To each of the fuel cells EX2a to EX2r using the separators of the present invention and above-mentioned comparative fuel cell CE1 was supplied a humidified, simulated fuel gas containing 70 volume % of H<sub>2</sub>,  
15 20 volume % of CO<sub>2</sub> and 10 volume % of H<sub>2</sub>O through the flow channels on the anode side, and supplied air as an oxidizing agent through the flow channels on the cathode side, so that each of the fuel cells not only EX2a to EX2r but also CE1 and CE4 was evaluated with respect to the stability of electric generation. Material used for conductive film, initial electric  
20 generation voltage, electric generation voltage after working for 10 days, and condition of separator after working for 10 days of each fuel cell were shown in Table 2.

[0033]

Table 2

Fuel cell	Material used for conductive film	Initial electric generation voltage (V)	Electric generation voltage after working for 10 days (V)	Condition of separator after working for 10 days
EX2a	Pt	0.78	0.77	a*
EX2b	Au	0.82	0.81	a*
EX2c	Pd	0.77	0.75	a*
EX2d	Ru	0.71	0.69	a*
EX2e	Rh	0.75	0.71	a*
EX2f	Ir	0.79	0.78	a*
EX2g	Ag	0.77	0.71	a*
EX2h	Pt-Au (10 weight %)	0.78	0.77	a*
EX2i	Pt-Pd (10 weight %)	0.78	0.74	a*
EX2j	Pt-Ru (5 weight %)	0.76	0.73	a*
EX2k	Pt-Rh (5 weight %)	0.75	0.72	a*
EX2l	Pt-Ir (30 weight %)	0.79	0.75	a*
EX2m	Au-Pd (30 weight %)	0.75	0.74	a*
EX2n	Au-Ir (30 weight %)	0.69	0.66	a*
EX2o	Au-Ag (50 weight %)	0.74	0.71	a*
EX2p	Pd-Ag (50 weight %)	0.76	0.70	a*
EX2q	Carbon	0.82	0.81	a*
EX2r	SiC	0.84	0.83	a*
CE1	Graphite	0.84	0.83	—
CE4	Al	0.47	0.21	d*

a\*: Corrosion was not observed.

d\*: Corrosion was observed in whole surface of flow channels.

5

[0034]

As shown in Table 2, the separators of the present invention each having an above-described, preferred conductive film were further excellent in the corrosion resistance, whereby the fuel cells EX2a to EX2r using the separators exhibited higher stability of electric generation.

10

[0035]

Example 3

Fuel cells EX3a to EX3j using the separators of the present invention were produced in the same manner as the above-mentioned fuel cell EX1 except that the porosity of the anodized aluminum layer was changed as shown in Table 3 by controlling conditions for disposing the anodized aluminum layer, respectively. Each of the fuel cells EX3a to

15

EX3j was evaluated with respect to the stability of electric generation in the same manner as Example 2. Porosity of anodized aluminum layer, initial electric generation voltage, electric generation voltage after working for 10 days, and condition of separator after working for 10 days of each fuel cell were shown in Table 3.

[0036]

Table 3

Fuel cell	Porosity of anodized aluminum layer (%)	Initial electric generation voltage (V)	Electric generation voltage after working for 10 days (V)	Condition of separator after working for 10 days
EX3a	0.86	0.81	0.83	a*
EX3b	1.25	0.82	0.81	a*
EX3c	1.63	0.86	0.84	a*
EX3d	2.01	0.83	0.81	a*
EX3e	2.67	0.79	0.75	b*
EX3f	3.02	0.84	0.78	b*
EX3g	3.54	0.85	0.75	b*
EX3h	5.06	0.88	0.73	b*
EX3i	7.32	0.81	0.63	c*
EX3j	10.55	0.86	0.59	c*

a\*: Corrosion was not observed.

b\*: Corrosion was scarcely observed.

c\*: Corrosion was observed in part of flow channels.

[0037]

As shown in Table 3, according to Example 3, the porosity of the anodized aluminum layer contained in the separator of the present invention was preferably 5.06 % or less, more preferably 2.01 % or less.

[0038]

#### Example 4

Fuel cells EX4a to EX4j using the separators of the present invention were produced in the same manner as the above-mentioned fuel cell EX1 except that the thickness of the anodized aluminum layer was changed as shown in Table 4 by controlling time for disposing the anodized aluminum layer, respectively. Incidentally, the anodized aluminum layer of each separator had a porosity of approximately 1.25 %. Each of the

fuel cells EX4a to EX4j was evaluated with respect to the stability of electric generation in the same manner as Example 2. Thickness of anodized aluminum layer, initial electric generation voltage, electric generation voltage after working for 10 days, and condition of separator after working for 10 days of each fuel cell were shown in Table 4.

[0039]

Table 4

Fuel cell	Thickness of anodized aluminum layer ( $\mu\text{m}$ )	Initial electric generation voltage (V)	Electric generation voltage after working for 10 days (V)	Condition of separator after working for 10 days
EX4a	3.5	0.86	0.71	c*
EX4b	4.8	0.83	0.81	b*
EX4c	9.7	0.82	0.84	a*
EX4d	15.8	0.85	0.81	a*
EX4e	26.7	0.88	0.86	a*
EX4f	33.1	0.81	0.78	a*
EX4g	51.5	0.81	0.76	b*
EX4h	69.1	0.85	0.69	c*
EX4i	78.3	0.88	0.63	c*
EX4j	94.6	0.86	0.56	c*

a\*: Corrosion was not observed.

b\*: Corrosion was scarcely observed.

c\*: Corrosion was observed in part of flow channels.

[0040]

As shown in Table 4, according to Example 4, the thickness of the anodized aluminum layer contained in the separator of the present invention was preferably 4.8 to 51.5  $\mu\text{m}$ , more preferably 9.7 to 33.1  $\mu\text{m}$ .

[0041]

#### Example 5

Fuel cells EX5a to EX5g using the separators of the present invention were produced in the same manner as the above-mentioned fuel cell EX1 except that the porosity of the anodized aluminum layer was stepwise changed by controlling anodizing conditions to dispose the anodized aluminum layer composed of a dense anodized aluminum layer and a porous anodized aluminum layer having a porosity shown in Table 5 provided thereon, respectively. Incidentally, the dense anodized

aluminum layer had a thickness of 15  $\mu\text{m}$  and a porosity of 1.25 %, and the porous anodized aluminum layer had a thickness of approximately 10 to 30  $\mu\text{m}$ . Each of the fuel cells EX5a to EX5g was evaluated with respect to the stability of electric generation in the same manner as Example 2.

- 5 Porosity of porous anodized aluminum layer, initial electric generation voltage, electric generation voltage after working for 36 days, and condition of separator after working for 36 days of each fuel cell were shown in Table 5.

[0042]

10 Table 5

Fuel cell	Porosity of porous anodized aluminum layer (%)	Initial electric generation voltage (V)	Electric generation voltage after working for 36 days (V)	Condition of separator after working for 36 days
EX5a	5.6	0.79	0.67	c*
EX5b	9.4	0.82	0.78	b*
EX5c	19.4	0.86	0.83	a*
EX5d	26.1	0.85	0.82	a*
EX5e	36.7	0.84	0.82	a*
EX5f	39.4	0.80	0.77	a*
EX5g	46.2	0.83	0.79	a*

a\*: Corrosion was not observed.

b\*: Corrosion was scarcely observed.

c\*: Corrosion was observed in part of flow channels.

15 [0043]

As shown in Table 5, according to Example 5, the porosity of the porous anodized aluminum layer was preferably 9.4 % or more, more preferably 19.4 % or more in the case where the anodized aluminum layer of the separator was composed of the dense anodized aluminum layer and the porous anodized aluminum layer. In the separators used for the fuel cells 5a and 5b, corrosion was generated around crack of the anodized aluminum layer.

[0044]

#### Example 6

25 Fuel cells EX6a to EX6g using the separators of the present invention were produced in the same manner as the above-mentioned fuel



cell EX1 except that a corner portion formed between surfaces of the flow channel was shaped into a curved surface having a curvature radius shown in Table 6, respectively. Incidentally, the anodized aluminum layer had a thickness of approximately 15  $\mu\text{m}$  and a porosity of 1.25 %. Each of the fuel cells EX6a to EX6g was evaluated with respect to the stability of electric generation in the same manner as Example 2. The above curvature radius of the corner portion formed between surfaces of the flow channel, and condition of separator after working for 34 days of each fuel cell were shown in Table 6.

[0045]

Table 6

Fuel cell	Curvature radius of corner portion formed between surfaces of flow channel (mm)	Condition of separator after working for 34 days
EX6a	0.1	c*
EX6b	0.2	c*
EX6c	0.3	c*
EX6d	0.5	b*
EX6e	0.7	b*
EX6f	1.0	a*
EX6g	1.5	a*

a\*: Corrosion was not observed.

b\*: Corrosion was scarcely observed.

c\*: Corrosion was observed in part of flow channels.

[0046]

As shown in Table 6, according to Example 6, the above-described curvature radius of the corner portion formed between surfaces of the flow channel in the separator was preferably 0.5 mm or more, more preferably 1.0 mm or more. In the separators used for the fuel cells EX6a to EX6c, corrosion was generated around crack of the anodized aluminum layer, which was mainly formed at the corner portion formed between surfaces of the flow channel.

[0047]

Example 7

Fuel cells EX7a to EX7g using the separators of the present invention were produced in the same manner as the above-mentioned fuel cell EX1 except that a corner portion formed between a side surface of the flow channel and the contact face was shaped into a curved surface having a curvature radius shown in Table 7, respectively. Incidentally, a corner portion formed between surfaces of the flow channel was shaped into a curved surface having a curvature radius of 1.0 mm. Further, the anodized aluminum layer had a thickness of approximately 15  $\mu\text{m}$  and a porosity of 1.25 %. Each of the fuel cells EX7a to EX7g was evaluated with respect to the stability of electric generation in the same manner as Example 2. Curvature radius of the corner portion formed between the side surface of the flow channel and the contact face, initial electric generation voltage, electric generation voltage after working for 34 days, and condition of separator after working for 34 days of each fuel cell were shown in Table 7.

[0048]

Table 7

Fuel cell	Curvature radius of corner portion formed between side surface of flow channel and contact face (mm)	Initial electric generation voltage (V)	Electric generation voltage after working for 34 days (V)	Condition of separator after working for 34 days
EX7a	0.1	0.80	0.67	c*
EX7b	0.2	0.81	0.72	c*
EX7c	0.3	0.80	0.78	b*
EX7d	0.5	0.78	0.70	a*
EX7e	0.7	0.83	0.81	a*
EX7f	1.0	0.80	0.78	a*
EX7g	1.5	0.82	0.79	a*

a\*: Corrosion was not observed.

b\*: Corrosion was scarcely observed.

c\*: Corrosion was observed in part of flow channels.

[0049]

As shown in Table 7, according to Example 7, the above-described curvature radius of the corner portion formed between the side surface of the flow channel and the contact face in the separator was preferably 0.3

mm or more, more preferably 0.5 mm or more. In the separators used for the fuel cells EX7a and EX7b, corrosion was generated around crack of the anodized aluminum layer, which was mainly formed at the corner portion formed between the side surface of the flow channel and the contact face.

5 [0050]

### Example 8

Fuel cells EX8a to EX8f using the separators of the present invention were produced in the same manner as the above-mentioned fuel cell EX1 except for changing the purity of the aluminum metal plate as shown in Table 8, respectively. Each of the fuel cells EX8a to EX8f was evaluated with respect to the stability of electric generation in the same manner as Example 1. Purity of aluminum metal plate, initial electric generation voltage, electric generation voltage after working for 12 days, and condition of separator after working for 12 days of each fuel cell were shown in Table 8.

[0051]

Table 8

Fuel cell	Purity of aluminum metal plate (%)	Initial electric generation voltage (V)	Electric generation voltage after working for 12 days (V)	Condition of separator after working for 12 days
EX8a	99.23	0.78	0.63	c*
EX8b	99.50	0.81	0.74	b*
EX8c	99.76	0.79	0.74	b*
EX8d	99.90	0.78	0.77	a*
EX8e	99.97	0.83	0.81	a*
EX8f	99.99	0.85	0.83	a*

a\*: Corrosion was not observed.

b\*: Corrosion was scarcely observed.

c\*: Corrosion was observed in part of flow channels.

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[0052]

As shown in Table 8, according to Example 8, the purity of the aluminum metal plate used for the separator of the present invention was preferably 99.50 % or more, more preferably 99.90 % or more.

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[0053]

### **Effects of the Invention**

As described in detail above, a separator for a fuel cell the present invention is mainly composed of aluminum, to be high in mass-productivity and to be produced with a reduced cost as compared with the conventional separator composed of graphite. Moreover, in the present invention, there is formed partly an anodized aluminum layer on the substrate, thereby making it possible to obtain a separator composed of aluminum excellent in corrosion resistance. A fuel cell using the separator of the present invention exhibits high stability of electric generation.

### **BRIEF DESCRIPTION OF THE DRAWINGS**

**Fig. 1** is a schematic view showing an example of a fuel cell using a separator of the present invention;

**Fig. 2** is a schematic view showing an embodiment of a separator for a fuel cell as well as a fragmentary enlarged view showing a structure of an anodized aluminum layer thereof of the present invention;

**Fig. 3** is a fragmentary schematic view showing an example of a shape of a flow channel for reaction gas according to a separator for a fuel cell of the present invention; and

**Fig. 4** is a fragmentary schematic view showing a flow channel and contact faces that come into contact with electrodes or collectors according to a separator of the present invention.

### **Descriptions of Letters and Numerals**

1···Unit Cell,

2···Solid Electrolyte,

3···Anode,

- 4···Cathode,
- 5···Separator,
- 6···Anodized Aluminum Layer,
- 7···Conductive Film,
- 5 8, 9···Flow Channel for Reaction Gas,
- 10···Porous Anodized Aluminum Layer, and
- 11···Dense Anodized Aluminum Layer.

**ABSTRACT**

**Problems to be solved by the Invention:**

To provide a separator composed of aluminum for a fuel cell  
5 excellent in corrosion resistance.

**Solution:**

A separator for a fuel cell composed of an aluminum metal plate  
having flow channels and contact faces that come into contact with  
10 electrodes or collectors, wherein an anodized aluminum layer is disposed  
on each of the flow channels.

**Selected Drawings:**

Fig. 1

Figure 1 is a schematic cross-sectional view of a multi-layered structure. The structure consists of alternating layers of material 1 (hatched) and material 2 (dotted). Material 1 contains rectangular openings 3 and 4. Material 2 contains rectangular openings 5 and 6. The structure is divided into two main sections by a vertical line, with a bracket labeled 1 indicating the repeating unit. The openings are labeled 8 and 9.

Fig. 2

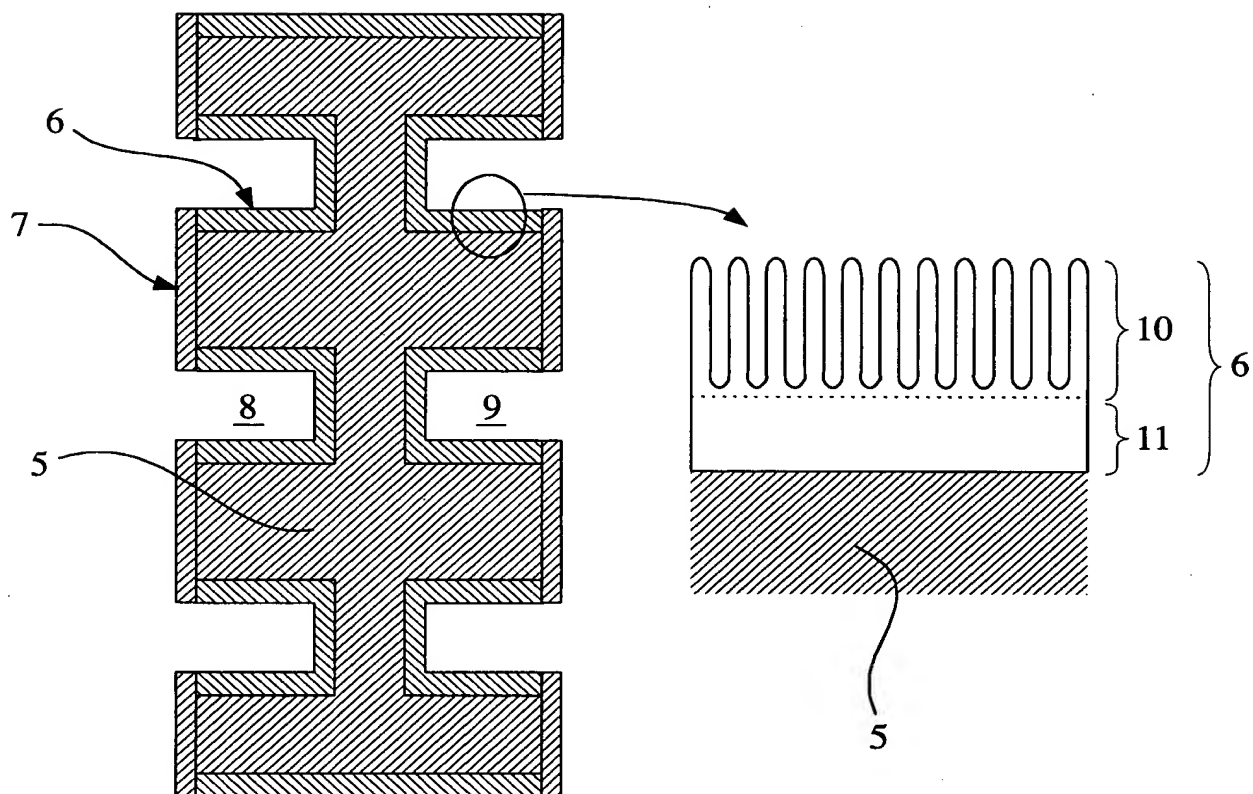




Fig. 3

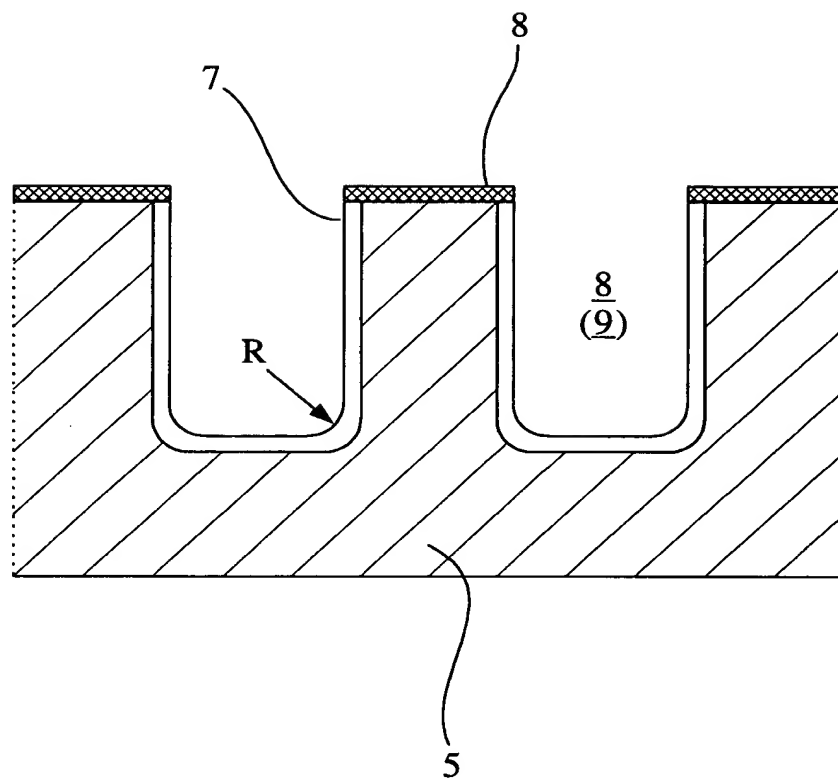


Fig. 4

